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## USE OF RUBBER CRUMB IN THE PRODUCTION OF BUILDING CERAMIC

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The prospects for using rubber crumb, obtained by processing automobile tires, in the production of building ceramic are examined. It is established on the basis of experimental data and preliminary statistical calculations that the addition of rubber crumb in amounts 5-10% changes the values of the physical–mechanical properties and structural indices of ceramic samples. The composition and amount of gas released during heat-treatment of ceramic samples is analyzed. It is established that additions have a negligible effect on the phase composition of ceramic samples. Rubber crumb makes it possible to obtain samples with satisfactory properties and to reclaim polymer wastes.

Key words: building ceramic, polymer wastes, rubber powder.

Wastes, technogenic raw materials, and industrial wastes, none of which have been previously used, are now used increasingly more often, together with the conventional raw materials, in the production of building materials [1-3].

The problem of reclaiming polymer wastes, such as worn tires, is topical, since because of their high resistance to destructive factor in nature such wastes are a source of environmental pollution. Dumps of worn tires occupy enormous amounts space and are a fire hazard. In many countries the reclamation of automobile tires is a serious environmental and economic problem.

In world practice, various methods are used to reprocess and recycle worn rubber. Rubber raw materials which are comminuted and separated from cord are used in the production of cement clinker and concrete, roofing materials, and new automobile tires, in road building, as fuel in heat-and-electricity plants, and so forth [4-6].

The objective of the present work is to study the possibility of using comminuted rubber crumb in the production of building ceramic. As result, it is possible to decrease the amount of such wastes and to increase the porosity of the ceramic.

Two types of illite-containing clays (clay 1 and clay 2), grog additions (sand and fireclay), and rubber crumb are used as the main raw materials for producing ceramic mix. The sintering interval of the clays used is small —  $30-70^{\circ}$ C. X-ray phase analysis has shown the presence of the following components in them: hydromica, chlorite, kaolinite, quartz, feldspars, dolomite, calcite, and hematite. The chemical composition of the initial clayey components is presented in Table 1.

Samples of clay 1 fired at temperature 950°C have average density 1.7 g/cm<sup>3</sup> and water absorption 16%. For clay 2 the analogous values are 2.0 g/cm<sup>3</sup> and 7.5%.

Sand was chosen as the grog material. X-ray phase analysis of the sand showed that its crystal structure consists of quartz, feldspars, and dolomite. The particle-size composition of the sand is represented by fine particles smaller than 2.5 mm with particles smaller than 0.315 mm predominating

**TABLE 1.** Chemical Composition of Clay

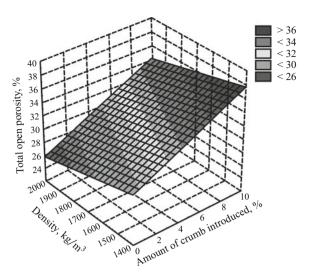
Raw material	Content, wt.%								
	$\mathrm{SiO}_2$	$Al_2O_3$	$Fe_2O_3$	CaO	MgO	MnO	$K_2O$	Na <sub>2</sub> O	other
Clay 1	47.60	17.82	7.66	6.27	3.60	0.09	4.50	0.55	11.50
Clay 2	66.33	15.80	6.42	1.80	2.72	_	1.63	_	5.30

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**Fig. 1.** Effect of the amount of rubber crumb introduced into the molding mix on the density and total open porosity of the ceramic samples.

(70% on the whole). The rubber crumb consists of 0.1 - 3.0 mm diameter granules.

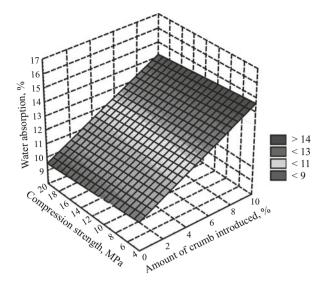
The compositions of the molding mixes are presented in Table 2.

The laboratory samples consisting of small cubes with 70 mm edges for determining the main properties and structural indices and 50 mm edges for determining compression strength as well as  $32 \times 40$  mm cylinders for analyzing the gas released during firing were produced by the plastic method. The samples were dried under natural conditions and finish-dried in a desiccator at  $100 - 105^{\circ}$ C. The firing temperature of the molding mixes was  $950^{\circ}$ C with isothermal soaking for 1 h. The temperature was raised to  $420^{\circ}$ C at a rate of 30 - 40 K/h and to  $950^{\circ}$ C at 80 - 90 K/h. The slow rate of temperature increase to  $420^{\circ}$ C was established experimentally; at higher rates macrostructural defects (deep cracks) formed on some samples while other samples were prone to swelling and deformation.

The physical-mechanical properties (total shrinkage, EN 772-13:2000 density, EN 772-1:2000 compression strength), operational properties (LST 1985:2006 cold-resistance), and structural indices (water absorption with soaking

**TABLE 2.** Ceramic Mix Compositions

26.11	Cor	%	
Molding mix No.	Clay mixtures 1 and 2	Grog additive (sand)	Rubber crumb
1	75	20	5
2	70	20	10
3	90	0	10
4	80	20	0



**Fig. 2.** Effect of the amount of rubber crumb introduced into the molding mix on the compression strength and on the water absorption of the ceramic samples.

for 72 h, total open porosity, degree of structural non-uniformity according to [7]) of the samples were determined.

During firing of the 32×40 mm cylindrical samples the composition of the released gases was analyzed. The carbon monoxide concentration was determined without feeding air.

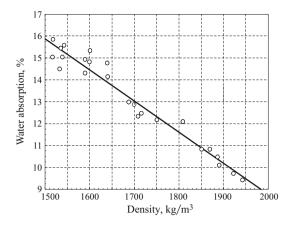
The average values of the corresponding indices of the samples are presented in Table 3.

The data presented show that when rubber crumb is introduced into the molding mix and with subsequent firing the density and compression strength of the samples decrease while the total shrinkage, water absorption, and total open porosity increase. In addition, the pores and capillaries in the samples gradually increase in size. The data in Table 3 show that the samples made with the molding mix 3 have the lowest density 1.57 g/cm³, the compression strength 10.2 MPa, and high shrinkage 16%.

It should be noted that all results obtained for the physical–chemical and structural indices were subjected to preliminary statistical analysis. The tendency of the content of the rubber crumb introduced into the molding mix to affect the density and total open porosity of the samples is shown in Fig. 1 while the affect on the compression strength and water absorption of the samples is shown in Fig. 2.

**TABLE 3.** Properties of the Samples after Firing at 950°C

Molding mix No.	Total shrinkage, %	Density, g/cm <sup>3</sup>	Compression strength, MPa	Water absorption, %	Total open porosity, %
1	12.3	1.70	6.5	12.4	33.0
2	9.3	1.60	6.3	15.1	36.8
3	16.0	1.57	10.2	14.8	36.4
4	6.9	1.89	15.0	10.2	26.5



**Fig. 3.** Density dependence of the water absorption of the ceramic samples.

The density dependence of the water absorption of the ceramic samples is shown in Fig. 3.

The density dependence of the water absorption of the ceramic samples can be described by the following empirical regression equation (with determination coefficient R = 0.954):

$$W = 37.23 - 0.014\rho$$
,

where W is the water absorption, in %, and  $\tilde{n}$  is the density, in  $kg/m^3$ .

Samples made with the molding mix 3 were used to analyze the composition and to determine the amount of gas released during firing (Fig. 4).

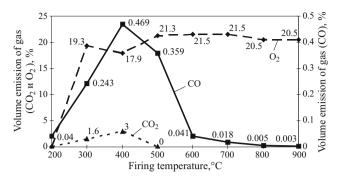
It is evident from the data that carbon monoxide release starts at firing temperature 200°C and that this process continues to temperature 900°C. The maximum carbon monoxide release was determined at 400°C and equals 0.469%. It is known [8] that when 8% sawdust is introduced into the molding mix the maximum carbon monoxide release is of the order of 2%, i.e., four times higher than the release obtaining when experimental polymer additive is introduced into the molding mix.

It is well known that incomplete combustion of organic additives results in the formation of carbon monoxide, which in air converts into carbon dioxide quite rapidly, so that the maximum carbon dioxide release 3% is established at temperature 400°C.

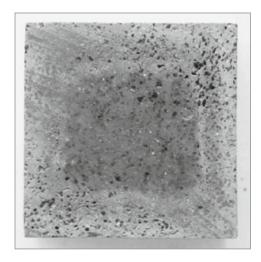
Smoke is released intensively at temperature about  $340^{\circ}$ C. Smoking decreases as the firing temperature increases to  $500^{\circ}$ C and stops completely at  $520^{\circ}$ C. The maximum oxygen release occurs at temperature  $600-700^{\circ}$ C.

It was also determined that the experimental additive as an organic substance forms a black center in the burnt samples (Fig. 5).

In our opinion, the black center is formed in the samples because during firing CO, H<sub>2</sub>, and other gases are released from the rubber crumb together with water vapor, which impedes oxygen penetration to the center of the samples. Thus, firing of the inner part of the samples occurs in a reducing



**Fig. 4.** Analysis of the composition and amount of gas released during firing of the samples made with the molding mix 3.



**Fig. 5.** Section of a burnt sample made from the molding mix 2.

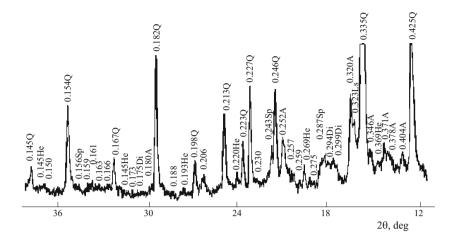
medium while firing of the surface of the samples occurs in an oxidizing medium, and as a result the surface of the samples is light brown in color. The transition  $Fe^{2+} \rightleftharpoons Fe^{3+}$  in the firing medium also affects the formation of a black center. The volume of the black center changes as rubber crumb and grog additive are introduced into the molding mix. If only rubber crumb is used (molding mix 3), then the volume of the black center is considerably larger than when rubber crumb and a grog additive are used together (molding mix 2).

Many authors believe that when such consumable additives are introduced into the mix a nonuniform structure of pores and capillaries is formed in the samples during firing, and this can have a negative effect on the cold-resistance of the samples. The additive halves the degree of structural nonuniformity of the samples.

It should be noted that the experimental samples with 10 wt.% rubber crumb added had a larger number of exterior pores, whose average diameter was 700  $\mu m$ . The cold-resistance of the experimental ceramic samples was >50 cycles with trilateral freezing and >75 cycles with unilateral freezing.

The phase composition of the samples was also determined in these investigations. The following crystalline

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**Fig. 6.** X-ray diffraction pattern of a fired sample made with molding mix 1.

phases were found in the x-ray diffraction pattern (Fig. 6) of the fired samples: quartz (Q 0.425, 0.335, 0.246, 0.227, 0.223, 0.213, 0.198, 0.182, 0.167, 0.154, 0.145 nm), anortite (A 0.404, 0.378, 0.371, 0.346, 0.320, 0.180 nm), diopside (Di 0.299, 0.294 nm), gehlenite (G 0.287 nm), hematite (He 0.369, 0.269, 0.220, 0.193, 0.164, 0.148 nm), and spinel (Sp 0.287, 0.243, 0.156 nm). This has a negligible effect on the phase composition of other molding mixes, including that of a mix with no rubber crumb.

In summary, these experimental studies have shown that 5-10% rubber crumb added to the molding mix increases the total open porosity of the samples by 6-10% as a result of the crumb being burned up. In turn, this increase of the porosity decreases the density of the samples. The rubber crumb, being the main source of the carbon monoxide release, whose maximum amount forms at the stage where the molding mixes are fired, does not make any qualitative changes in the phase composition of the material obtained and can be used as a consumable additive in the production of ceramic building materials.

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